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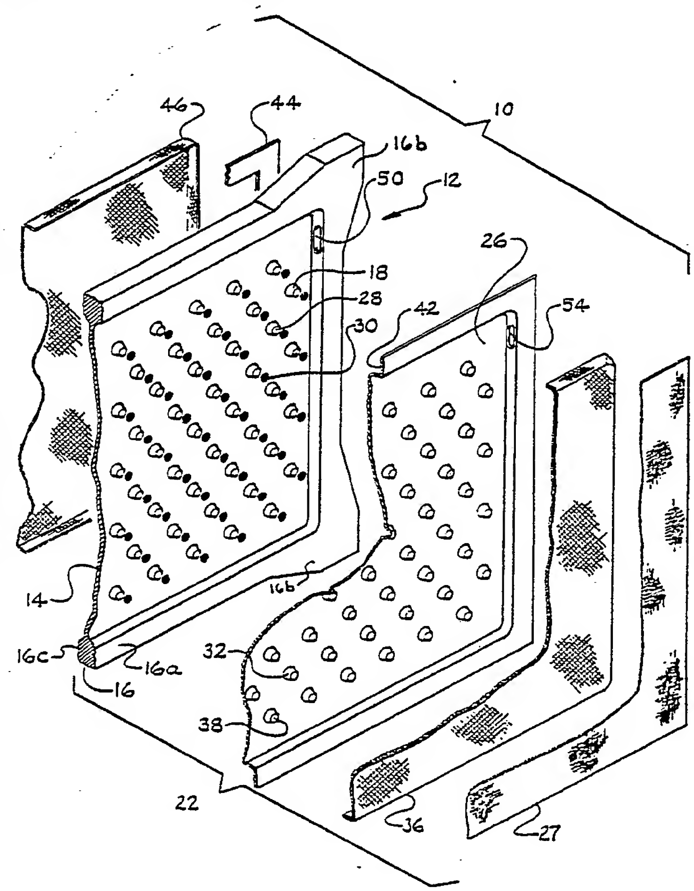
(54) Title: UNITARY CENTRAL CELL ELEMENT FOR FILTER PRESS ELECTROLYSIS CELL STRUCTURE

(57) Abstract

Unitary, cast structural element (10) for a filter press electrolysis cell which incorporates into a single unit the central barrier (14) between the peripheral boundaries for the adjacent anolyte compartment (22) and the adjacent catholyte compartment (24) of two electrolysis cells located on opposite sides of the central barrier (14). Also incorporated into the single cast structural element (10) are anode bosses (18) and cathode bosses (20) extending outwardly from opposite sides of the central barrier. These bosses (18, 20) not only serve as mechanical support for their respective flat plate anode (36) and cathode (46), but also serve as stand-off means and electrical current collectors and disperses from the cathode (46) of one electrolysis cell to the anode (36) of the next cell. Simplicity of design coupled with incorporation of many functional elements into one part eliminates many warpage problems, inherent high voltage problems and membrane (27) 'hot spot' problems.



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UNITARY CENTRAL CELL ELEMENT FOR FILTER PRESS ELECTROLYSIS CELL STRUCTURE

This invention relates to an improvement in the structure of bipolar electrode-type, filter press--type electrolysis cells. More particularly, it relates to those of such cells which employ permselective ion exchange membranes which are disposed in a plane between flat surfaced, parallel, foraminous, metal anodes and cathodes when said anodes and cathodes are mounted at a distance from the fluid impermeable structure of the bipolar electrode which physically separates adjacent electrolysis cells. Such cells are particularly useful in the electrolysis of aqueous solutions of alkali metal chlorides; especially in the electrolysis of aqueous solutions of sodium chloride (brine). The cell structure may also be used in electrolyzing other solutions to make products such as potassium hydroxide, iodine, bromine, bromic acid, persulfuric acid, chloric acid, deuterium, tritium, adiponitrile and other organic compounds made by electrolysis.

The unitary filter press central cell element of the present invention decreases the cost of

manufacture of the cell units, decreases the labor required to assemble them, simplifies their manufacture, greatly reduces the warpage of the cell unit parts and provides a much sturdier cell structure than do bipolar, filter press cells of the prior art.

Reducing the warpage of cell structure allows the cell to be operated more efficiently; i.e., produce more units of electrolysis products per unit of electricity. Reducing the warpage reduces the deviation from design of the gap width between the anode and cathode of each electrolysis cell. Ideally this gap width is uniformly the same between the anode and cathode in order to have a uniform current density spread between the faces of the cell electrodes. Among other things, structural warpage causes deviation of this gap resulting in some parts of the anode and cathode being closer together than others. At these locations, the electrical resistance is less, the electrical current flow is higher and, thus, the electrical heating is greater. This electrical heating is sufficient in many instances to cause damage to the membrane at these locations. These locations of unacceptably high electrical current concentration and high heat are referred to herein as "hot spots".

To avoid these hot spots, the prior art has had to design its cell structures with a greater than desired gap width between the anode and cathode of each electrolysis cell. This, of course, increases the cell operating voltage and decreases the cell operating efficiency. Complexity of design and fabrication is another drawback of those cells.

Examples of bipolar filter press cells operated in a cell series are disclosed in Seko, U.S. Patent No. 4,111,779 (September 5, 1978) and in Pohto, U.S. Patent No. 4,017,375 (April 12, 1977). Other representative flat plate bipolar electrode-type, filter press-type electrolytic cells can also be observed in U.S. Patent Nos. 4,364,815; 4,115,236; 3,960,698; 3,859,197; 3,752,757; 4,194,670; 3,788,966; 3,884,781; 4,137,144 and 3,960,699.

The prior art also discloses complex and elaborate schemes devised to electrically and/or mechanically connect different parts of the cell wherein titanium and titanium alloys are employed. Particularly is this complexity seen to be true with respect to the parts herein referred to as stand-offs which connect the "flat plate" anode and cathode of a bipolar electrode structure to an electrically conductive central barrier at a spaced distance from the central barrier; e.g., see U.S. Patent Nos. 4,111,779 and 4,194,670. Other stand-offs are used to support the flat plate electrodes and to electrically and mechanically connect them through holes in a non--conductive central barrier, e.g., see U.S. Patent Nos. 3,752,757 and 3,960,698. It will be noticed that in these connections, welds and/or bolts are used to connect the stand-offs to the electrodes and then again to the central barrier or to opposing stand-offs passing through the central barrier. Many problems are associated with these connections in order to get adequate electrical current distribution.

The present invention reduces these problems by eliminating many of these connections. It does

this by integrally casting these stand-offs with the central barrier. Moreover, the connections used to connect the central barrier to the peripheral structure of the anolyte and catholyte compartment are also eliminated by integrally casting these structures with the central barrier.

The present invention by comparison (cast unitary cell structures) has eliminated most of the problems which are common to the weldment-type structure and the welded and bolted structure. As a result, cell electrodes are more uniformly parallel; there is a more uniform distribution of electrical current and electrolytic reaction in the cell during operation; and the invention also provides a leakproof centerboard or central barrier.

The present invention also greatly reduces the risk of titanium hydride formation by creating a structure which has a titanium liner with only a relatively very few stress points in it, and also by locating these stress points at an extreme distance from the hydrogen source with respect to the amount of steel which must be traversed in order to reach any of these few stress points. The only stress points found in the present invention's titanium hot pressed liner are found at the sites where it is welded to the ends of the integrally cast anode bosses. These will be discussed below. It should be understood here, however, that although the present invention has been discussed principally in terms of the commonly used steel and titanium, it is not limited to these materials of construction, albeit they are the preferred material of construction.

The present invention more particularly resides in a cell structure used in forming a bipolar electrode-type, filter press-type electrolytic cell unit. This particular cell unit is capable of being combined with other cell units to form a cell series. In the cell series, the cell structure is separated from adjacent cell structures by ion-exchange, perm-selective membranes which are sealably disposed between each of the cell structures so as to form a plurality of electrolysis cells. Each of said electrolysis cells has at least one planarly disposed membrane defining and separating the anolyte compartment from the catholyte compartment of each electrolysis cell. The cell structure of this particular cell unit has a central barrier which physically separates the anolyte compartment of a cell located on one side of the barrier from the catholyte compartment of an adjacent cell located on the opposite side of the barrier. This central barrier has a planarly disposed foraminous, "flat plate" anode situated in its adjacent anolyte compartment and a planarly disposed, foraminous, "flat plate" cathode situated in its adjacent catholyte compartment. Both electrode faces are substantially parallel to the membrane planarly disposed between them and to the central barrier. The central barrier has the anode of the adjacent anolyte compartment electrically connected through it to the cathode of the adjacent catholyte compartment.

These anolyte and catholyte compartments adjacent the central barrier have a structure around their periphery to complete their physical definition. This cell structure also has an electrical current

transfer means associated with it for providing electrical current passage through the central barrier from its adjacent catholyte compartment to its adjacent anolyte compartment. This cell structure includes anode and cathode stand-off means for maintaining the anode and cathode of the two electrolysis cells adjacent the central barrier at predetermined distances from the central barrier.

The improvement of this particular cell structure comprises the central barrier, the anolyte and catholyte compartment peripheral structures, the anode stand-off means, the cathode stand-off means, and at least part of the electrical current transfer means all being integrally formed into a unitary central cell element made from a single casting of a castable metal.

The invention employs the castable metal as part of the electrical current transfer means which transfers electricity through the central barrier from the adjacent catholyte compartment to the adjacent anolyte compartment.

The unitary central cell element is formed in a fashion so as to provide the structural integrity required to physically support the adjacent electrolyte compartments while loaded with electrolyte as well as to support the associated electrolysis cell appurtances which are desired to be supported by the unitary central cell element.

The anode stand-off means and that part of the electrical current connecting means located in

the unitary central cell element on the anolyte side of the central barrier are combined into a multiplicity of anode bosses projecting a predetermined distance outwardly from the central barrier into the anolyte compartment adjacent the central barrier. These anode bosses are capable of being mechanically and electrically connected either directly to the anode of said anolyte compartment or indirectly to said anode through at least one compatible metal intermediate directly situated in an abutting fashion between said anode and said anode bosses. Preferably, these anode bosses all have ends which are flat surfaces which preferably lie in the same geometrical plane.

The cathode stand-off means and that part of the electrical current connecting means located on the catholyte side of the central barrier are combined into a multiplicity of cathode bosses projecting a predetermined distance outwardly from the central barrier. These cathode bosses are capable of being mechanically and electrically connected either directly to the cathode in said adjacent catholyte compartment or indirectly to the cathode through at least one weldably compatible metal intermediate directly situated in an abutting fashion between said cathode and said cathode bosses. Preferably these cathode bosses all have ends which are flat surfaces and which preferably lie in the same geometric plane.

The invention preferably further comprises anode bosses being spaced apart in a fashion such that anolyte can freely circulate through the totality of the otherwise unoccupied adjacent anolyte compartment and, likewise, said cathode bosses being spaced

apart in a fashion such that catholyte can freely circulate throughout the totality of the otherwise unoccupied adjacent catholyte compartment.

Preferably, the castable material of the unitary central cell element is selected from iron, steel, stainless steel, nickel, aluminum, copper, chromium, magnesium, tantalum, cadmium, zirconium, lead, zinc, vanadium, tungsten, iridium, rhodium, cobalt, alloys of each, and alloys thereof.

More preferably, the metal of the unitary cell element is selected from ferrous materials. Ferrous materials are defined herein to mean metallic materials whose primary constituent is iron.

A further element which this invention preferably includes is an anolyte side liner made of a metal sheet fitted over those surfaces on the anolyte compartment side of the cell structure which would otherwise be exposed to the corrosive environment of the anolyte compartments.

Preferably, this anolyte side liner is an electrically conductive metal which is essentially resistant to corrosion due to the anolyte compartment environment, and which is formed so as to fit over and around the anode bosses with the liner being connected to the unitary central cell element at the anode bosses more preferably connected at the ends of the anode bosses.

Preferably, the invention also comprises having the liner sufficiently depressed around the

spaced anode bosses toward the central barrier in the spaces between the bosses so as to allow free circulation of the anolyte between the lined unitary central cell element and the membrane of the adjacent anolyte chamber. Note that the liner replaces the unitary central cell element surface adjacent to the anolyte chamber as one boundary contacting the anolyte.

More preferably, the metal liner is connected to the anode bosses by welding through a metal intermediate which is disposed between the bosses and the liner with the metal of the metal intermediate being weldably compatible with both the metal of the anolyte side liner and the metal of which the unitary central cell element is made, that is weldably compatible with both metals to the point of being capable of forming a ductile solid solution with them at welds of them upon their welding.

In most cases, such as in the construction of chloralkali cells, it is preferred that the unitary cell element be made of a ferrous material and the anolyte side liner be made of a metallic material selected from titanium, titanium alloys, tantalum, tantalum alloys, niobium, niobium alloys, hafnium, hafnium alloys, zirconium and zirconium alloys.

In situations where the anolyte side liner metal is not weldably compatible with the metal of the unitary cell element, then in order to be able to weld the liner to the structure, metal coupons are one type of metal intermediate which are suitable to be situated in an abutting fashion between the anode bosses and the anolyte side liner. Each coupon has at least two metal

layers bonded together, with the outside metal layer of one side of the coupon abutting the anode boss and the outside metal layer of the opposite side of the coupon abutting the anolyte side liner. The metal layer of the coupons which abuts each anode boss is weldably compatible with the material of which the anode bosses are made and accordingly being welded to said anode bosses. The metal layer of that side of the coupons abutting the anolyte side liner is weldably compatible with the metallic material of which the anolyte side liner is made and accordingly is welded to said liner so that the liner is welded to the anode bosses through the coupons. In some instances, wafers made of a single metal or metal alloy serve quite well as intermediates.

In most cases, it is preferred that the anolyte side liner be made of titanium or a titanium alloy, and the castable material from which the unitary central cell element be made is a ferrous material.

In the situation where the anolyte liner is titanium material and the anode bosses are a ferrous material, then it is preferred to have vanadium wafers serve as the weldably compatible metal intermediates interposed between the anode bosses and the adjacent anolyte side liner so that the titanium anolyte side liner can be welded to the ferrous material anode bosses through the vanadium wafers. Vanadium is a metal which is weldably compatible with both titanium and ferrous material.

In some instances, it is preferred to have the metal intermediates situated between the anode

bosses and the adjacent anolyte side liner joined to the ends of the anode bosses by a film-forming process. Spraying a hot liquid metal, such as vanadium, is one film-forming process. Another film-forming process is carried out by soldering or brazing the metal to the anode bosses.

In some occasions it is found that no metal intermediate is required to be used between the liner and the anode bosses, and that the anolyte side liner can be directly bonded to the anode bosses by welding.

Another way of connecting an anolyte liner to the unitary cell structure when these metals are weldably incompatible is that where no metal intermediate is used, but wherein the anolyte side liner is bonded directly to the anode bosses by explosion bonding.

In many instances, it is desired that the anolyte side metal liner extend over the lateral face of the anolyte compartment peripheral structure so as to form a sealing face thereat for the membrane when the cell segments are squeezed together to form a cell series.

In most instances, it is desired that the anolyte side liner be connected to the unitary central cell element at the ends of the anode bosses. However, this invention includes connecting the liner to the sides of these bosses and even connecting the liner to the central barrier between the bosses. Preferably,

however, the anolyte side liner is welded to the ends of the anode bosses through an intermediate metal coupon or wafer.

A catholyte liner is usually required less frequently than an anolyte liner. However, there are many occasions, such as in high concentration caustic catholyte compartments, wherein a catholyte side liner is needed on the catholyte side of the unitary cell element. Thus this invention also comprises a catholyte side liner made of a metal sheet fitted over these surfaces of the unitary central cell element which would otherwise be exposed to the catholyte compartment of the adjacent electrolysis cell.

This catholyte side liner is made from an electrically conductive metal which is essentially resistant to corrosion due to the catholyte compartment environment. Plastic liners may be used in some cases where provision is made for electrically connecting the cathode to the cathode bosses through the plastic. Also combinations of plastic and metal liners may be used. The same is true for anolyte side liners.

The catholyte liner is depressed sufficiently around the spaced cathode bosses toward the central barrier in the spaces between the bosses so as to allow free circulation of the catholyte between the lined unitary central cell element and the membrane of the adjacent catholyte chamber. Note that the liner replaces the unitary central

cell element surface adjacent to the catholyte chamber as one boundary contacting the catholyte.

Unlike the anolyte side liner, it is preferred that the metal catholyte side liner be directly connected to the cathode bosses by welding without a metal intermediate being disposed between the bosses and the liner. A metal intermediate can be used, however. If so, then the metal intermediate must be weldably compatible with both the metal of of the catholyte side liner and the metal of which the unitary cell element is made.

In many instances it is desired that the unitary cell element be made of a ferrous material and the metal for the catholyte side liner be selected from ferrous materials, nickel, nickel alloys, chromium, magnesium, tantalium, cadmium, zirconium, lead, zinc, vanadium, tungsten, iridium, and cobalt.

In many instances it is desired that the metal of the unitary central cell element, of the catholyte side liner, and of the cathode of the adjacent electrolysis cell be all selected from ferrous materials.

In some instances it is preferred to have the metal intermediates situated between the cathode bosses and the adjacent catholyte side liner joined to the ends of the cathode bosses by a film-forming process. Spraying a hot liquid metal is one film-forming process. Another film--forming process is carried out by soldering or brazing the metal to the cathode bosses.

However, in most cases, the metal of the catholyte liner can be welded directly to the unitary cell structure without the need of metal intermediate. Nickel is usually the most preferred catholyte liner material.

The catholyte side metal liner is formed so as to fit over and around the ends of the cathode bosses and is welded directly on one side of the liner to the bosses in a manner so as to provide an electrical connection between the unitary central cell element and the cathode. The cathode itself is directly welded to the opposite side of the cathode side liner.

As with the anolyte side liner, it is preferred that the catholyte side metal liner also extend over the lateral face of the catholyte compartment peripheral structure so as to form a sealing face thereat for the membrane when the cell segments are squeezed together to form a cell series.

In most instances it is desired that the catholyte side liner be connected to the unitary central cell element at the ends of the cathode bosses. However, this invention includes connecting the liner to the central barrier between the bosses.

The invention can be better understood by reference to the drawing illustrating the preferred embodiment of the invention, and wherein like reference numerals refer to like parts in the different drawing figures, and wherein:

Fig. 1 is an exploded, partially broken-away perspective view of a unitary cell element 12 of this invention shown with accompanying parts forming one bipolar electrode type filter press-type cell unit 10 of a cell series of such cell units;

Fig. 2 is a cross-sectional side view of three filter press-type cell units 10 employing the unitary cell elements 12 of the present invention. The cell units are shown as they would appear in a filter press cell series. The cross-section is taken along and in the direction of line 2-2 in Figs. 4 and 5;

Fig. 3 is an exploded, sectional side view of a cell structure taken along line 3-3 in Figs. 4 and 5.

Fig. 4 is a partially broken-away front view of a cell unit 10 as viewed from the cathode side; and

Fig. 5 is a partially broken-away front view of a cell unit 10 as viewed from the anode side.

Referring to Figs. 1, 2 and 3, a bipolar electrode-type, filter press-type electrolysis cell unit 10 is shown employing the preferred embodiment of the unitary central cell element 12 of this invention. This cell is also referred to as a flat plate cell.

In the preferred embodiment, the cell element 12 is made of cast steel. It has a solid central barrier 14, a peripheral flange 16 extending laterally from both sides of the periphery of the central barrier 14, protruding and spaced-apart anode bosses 18, and protruding and spaced-apart cathode bosses 20.

By having these parts all integrally cast into one element 12, many problems are simultaneously eliminated or greatly reduced. For example, most of the warpage problems, fluid leakage problems, electric current maldistribution problems, and complications of cell construction on a mass production basis are greatly alleviated. This simplicity of cell design allows cell elements to be constructed which are much more reliable and which are constructed at a much more economical cost.

An anolyte compartment 22 of an adjacent cell can be seen on the right side of cell element 12. On the left side of cell element 12, a catholyte compartment 24 of a second adjacent cell can be seen. Thus, cell element 12 separates one cell from another. One very important feature in cells of this type is to get electricity from one cell to another as cheaply as possible.

On the anolyte compartment side of central element 12, there is a liner 26 made of a single sheet of titanium. This liner 26 is hot formed by a press in such a fashion so as to fit over and substantially against the surfaces of the unitary

central cell unit 12 on its anolyte compartment side. This is done to protect the steel of cell element 12 from the corrosive environment of the anolyte compartment 22. Liner 26 also forms the left boundary of anolyte compartment 22 with an ion-exchange membrane 27 forming the right boundary (as shown in Fig. 3). Cell element 12 is cast so that its peripheral structure forms the flange 16 which serves not only as the peripheral boundary of the anolyte compartment 22, but also as the peripheral boundary of the catholyte compartment 24. Preferably the titanium liner 26 is formed with no stresses in it in order to provide a liner which atomic hydrogen cannot attack as rapidly to form brittle, electrically nonconductive titanium hydrides. Atomic hydrogen is known to attack stressed titanium more rapidly. Avoiding these stresses in the liner is accomplished by hot forming the liner in a press at an elevated temperature of from 482-538°C. Both the liner metal and press are heated to this elevated temperature before pressing the liner into the desired shape. The liner is then held in the heated press for about forty-five minutes to prevent the formation of stresses in it as it cools to room temperature.

The titanium liner 26 is connected to the steel cell element 12 by resistance welding. This is accomplished indirectly by welding the liner 26 to the flat ends 28 of the frustoconically shaped, solid anode bosses 18 through vanadium wafers 30. Vanadium is a metal which is weldable itself and which is weldably compatible with titanium and steel. By weldably compatible is meant that

one weldable metal will form a ductile solid solution with another weldable metal upon the welding of the two metals together. Titanium and steel are not weldably compatible with each other, but both are weldably compatible with vanadium. Hence, the vanadium wafers 30 are used as an intermediate metal between the steel anode bosses 18 and the titanium liner 26 to accomplish the welding of them together to form an electrical connection between liner 26 and central cell element 12 as well as to form a mechanical support means for central cell element 12 to supporting liner 26.

The preferred fit of the anolyte side liner 26 against the central cell element 12 can be seen from the drawing (Fig. 2). The liner 26 has indentations or hollow caps 32 pressed into it. These caps 32 are frustoconically shaped and are hollow instead of being solid as are the anode bosses 18. The caps 32 are sized and spaced so that they fit over and around anode bosses 18. The depth of depression of the caps is such that their interior ends 34 abut the vanadium wafers 30 when the wafers 30 are abutting the flat ends 28 of anode bosses 18 and when these elements are welded together. The shape of the bosses and caps is not significant. They could be square shaped or any other convenient shape. However, their ends 28 should all be flat and should all lie in the same imaginary geometrical plane. In fact, these anode bosses and caps can be shaped and located so as to guide anolyte and gas circulation.

The liner 26 is resistance welded at the interior ends 34 of its caps 32 to the steel flat ends 28 of anode bosses 18 through the interposed, weldably compatible, vanadium wafers 30.

Anode 36 is a substantially flat sheet of expanded metal or woven wire made of titanium, preferably having a ruthenium oxide catalyst coating on it. It is welded directly to the outside of flat ends 38 of indented caps 32 of liner 26. These welds form an electrical connection and a mechanical support means for anode 36. Other catalyst coatings can be used.

In Fig. 2a, membrane 27 is disposed between the anode 36 of the one cell unit 10 and the cathode 46 of the next adjacent cell unit 10 so as to form an electrolysis cell between the central barrier 14 of each of the two adjacent unitary central cell elements 12.

Representative of the types of permselective membranes envisioned for use with this invention are those disclosed in the following U.S. Patents: 3,909,378; 4,329,435; 4,065,366; 4,116,888; 4,126,588; 4,209,635; 4,212,713; 4,251,333; 4,270,996; 4,123,336; 4,151,053; 4,176,215; 4,178,218; 4,340,680; 4,357,218; 4,025,405; 4,192,725; 4,330,654; 4,337,137; 4,337,211; 4,358, 412; and 4,358,547.

Of course, it is within the purview of this invention for the electrolysis cell formed between the two cell segments to be a multi-compartment electrolysis cell using more than one membrane,

e.g., a three-compartment cell with two membranes spaced from one another so as to form a compartment between them as well as the compartment formed on the opposite side of each membrane between each membrane and its respective adjacent cell unit 10.

The location of anode 36 within anolyte compartment 22 with respect to the membrane 27 and the titanium lined central barrier 14 is determined by the relationships between the lateral extension of flange 16 from central barrier 14, the extension of anode bosses 18 from the central barrier 14, the thickness of the vanadium wafers 30, the thickness of liner 26, and the like. It can be readily seen that anode 36 can be moved from a position abutting the membrane 27 to a position with some considerable gap between the membrane 27 and anode 36 by changing these relationships; e.g., changing the extension of anode bosses 18 from the central barrier 14. It is preferred, however, that the flange 16 on the anolyte side of central barrier 14 extend the same distance as do the anode bosses 18 from the central barrier 14. This adds to the simplification of construction of cell element 12 because, with this circumstance, a machine metal planar can plane both the end surfaces 28 of anode bosses 18 as well as the flange surface 16a at the same time in a manner so that these surfaces all lie in the same geometrical plane. The same preference is true for like surfaces on the catholyte side of cell element 12, i.e., it is preferred that the flat ends 40 of cathode bosses 20 and the flange surface 16c of flange 16 which lies on the catholyte side of element 12 all be machined so as to all lie in the same geometrical plane.

For fluid sealing purposes between membrane 27, and flange surface 16a, it is preferred for anolyte liner 26 to be formed in the shape of a pan with an off-set lip 42 extending around its periphery. Lip 42 fits flush against the flange face 16a. The periphery of membrane 27 fits flush against anolyte liner lip 42, and a peripheral gasket 44 fits flush against the other side of the periphery of membrane 27. In a cell series, as shown in Fig. 2, the gasket 44 fits flush against the flange face 16c on the catholyte side of the next adjacent cell element 12 and flush against membrane 27 when there is no liner 48.

Although only one gasket 44 is shown, this invention also encompasses the use of gaskets on both sides of membrane 27. It also encompasses the situation where no lip 42 is provided.

On the side of cell element 12 opposite the anolyte compartment side, i.e., the catholyte side, there is no catholyte side liner shown in Fig. 1, although there is a catholyte side liner 48 shown in Figs. 2, 3, and 4. This is done to illustrate the fact that the presence of two liners is sometimes desired but sometimes not. Most often the metal from which central cell element 12 is cast is also suitable for use in either the catholyte compartment 24 or the anolyte compartment 22. For example, in a cell series wherein aqueous solutions of sodium chloride are electrolyzed to form caustic and/or hydrogen gas in the catholyte compartment 24, then ferrous materials such as steel are quite suitable for the catholyte compartment metal

components at most cell operating temperatures and caustic concentrations, e.g., below about 22 percent caustic concentration and at cell operating temperatures below about 85°C. Hence, if cell element 12 is made of a ferrous material such as steel, and if caustic is produced at concentrations lower than about 22 percent and the cell is to be operated below about 85°C, then a protective liner is not needed on the catholyte side of cell element 12 to protect the steel of element 12 from corrosion. However, the titanium anolyte side liner 26 is still needed on the anolyte side. Hence, in Fig. 1, there is no catholyte side liner 48 shown. Instead, the flat foraminous metal cathode 46 (also made of steel in this embodiment in Fig. 1) is resistance welded directly to the ends 40 of the cathode bosses 20.

Referring to Figs. 2 and 3, the catholyte side (the left side) of cell element 12 is seen to appear as the mirror image of its anolyte side. The flange 16 forms the peripheral boundary of the catholyte compartment 24, while the central barrier 14 and membrane 27 form its remaining boundaries. Spaced cathode bosses 20 are solid, frustoconically--shaped protrusions extending outwardly from central barrier 14 into catholyte compartment 24. Flat--surfaced, foraminous, steel plate cathode 46 is welded directly to the flattened ends 40 of cathode bosses 20.

Referring to Figs. 2, 3, and 4, a catholyte side liner 48 made of a metal which is highly resistant to corrosive attack from the environment

of the catholyte compartment 24 is shown. The metal must also be sufficiently ductile and workable so as to be pressed from a single sheet of metal into the non-planar form shown. This includes being capable of having the indentations or caps 70 pressed into the sheet. The caps 70 are spaced so that they fit over and around the spaced cathode bosses 20. It is preferred that the catholyte side liner 48 have an indented lip 72 extending around its periphery in a fashion so as to abut the flange face 16c on the side of central cell element 12 which is adjacent the catholyte compartment 24. Liner 48 is preferably connected to central cell element 12 by resistance welding of the internal ends of the liner caps to the flat ends 40 of cathode bosses 20. That is, this is preferable if the metal of the liner 48 and the central support element 12 are weldably compatible with each other. If these metals are not weldably compatible, then there should be used metal wafers or intermediates or combinations of intermediates which are weldably compatible with the metals of liner 48 and cell element 12. These wafers 78 are disposed between the cathode boss flat ends 40 and the interior ends 74 of the liner caps 70 which correspond to the boss ends 40 and are welded to the ends 40 of cathode bosses 20. Catholyte liner 48 is then welded to the ends 40 of cathode bosses 20 through metal wafers 78. Cathode 46 is then welded to the external end 76 of caps 70. The connection of each liner cap 70 through a metal wafer 78 to the end 40 of a cathode boss 20 may be made with only one weld; i.e., the metal wafer does not have to be welded by itself beforehand.

Metal wafers 78 and 30 may be formed of a metal which is weldably compatible with both the metal of the cell element 12 and the metal of the respective liners 26 or 48. It should be noted that metal wafers can have more than two layers of metal such as a three layer explosion bonded wafer of titanium, copper and a ferrous material.

Both of the flat-surfaced anode 36 and the flat-surfaced cathode 46 have their peripheral edges rolled inwardly toward the cell element 12 and away from the membranes 27. This is done to prevent the sometimes jagged edges of these electrodes from contacting the membranes 27 and tearing it.

It should be noted that the corners 16b of central cell element peripheral flange 16 are built-up to allow the cell to be operated at higher pressures than atmospheric. Of course, the shape of the cell can be round as well as rectangular, or any other convenient shape. A round shape would probably be the most practical for very high pressure operations.

With brine as cell feed, the cell operates as follows. The feed brine is continuously fed into anolyte compartment 22 via duct 60 while fresh water may be fed into catholyte compartment 24 via duct 64 (Figs. 4 and 5). Electric power (D.C.) is applied across the cell series so that the anode 36 of each electrolysis cell is positive with respect to the cathode 46 of that electrolysis cell. Excluding depolarized cathodes or anodes, the

electrolysis proceeds as follows. Chlorine gas is continuously produced at the anode 36; sodium cations are transported through membrane 27 to the catholyte compartment by the electrostatic attraction of the cathode 46. In the catholyte compartment 24 there is hydrogen gas and an aqueous solution of sodium hydroxide continuously formed. The chlorine gas and depleted brine continuously flow from the anolyte chamber 22 via duct 62 while the hydrogen gas and sodium hydroxide continuously exit the catholyte compartment 24 by duct 66. Depolarized electrodes can be used to suppress the production of hydrogen or chlorine or both if desired.

In operating the cell series as an electrolysis cell series for NaCl brine, certain operating conditions are preferred. In the anolyte compartment a pH of from 0.5 to 5.0 is desired to be maintained. The feed brine preferably contains only minor amounts of multivalent cations (less than about 0.8 gram/liter when expressed as calcium). More multivalent cation concentration is tolerated with the same beneficial results if the feed brine contains carbon dioxide in concentrations lower than about 70 ppm when the pH of the feed brine is lower than 3.5. Operating temperatures can range from 0° to 250°C, but preferably above about 60°C. Brine purified from multivalent cations by ion-exchange resins after conventional brine treatment has occurred is particularly useful in prolonging the life of the membrane. A low iron content in the feed brine is desired to prolong the life of the membrane. Preferably the pH of

the brine feed is maintained at a pH below 4.0 by the addition of hydrochloric acid.

Preferably the pressure in the catholyte compartment is maintained at a pressure slightly greater than that in the anolyte compartment, but preferably at a pressure difference which is no greater than a head pressure of about 1 foot of water. Preferably this pressure difference is controlled by surge tanks such as is disclosed in U.S. Patent 4,105,515.

Preferably the operating pressure is maintained at less than 7 atmospheres.

Usually the cell is operated at a current density of from 1.0 to 4.0 amperes per square inch, but in some cases operating about 4.0 amps/inch is quite acceptable.

Where a metal liner is employed on both sides of the cell structure in a chlor-alkali cell, a catholyte side, liner 48 made of nickel is desirable when the caustic concentration in the catholyte compartment 24 is maintained above about 22 weight percent and the cell electrolyte operating temperature is maintained above about 80°C. This nickel liner 48 is formed, sized for, and fitted to the central cell element 12 in essentially the same manner as is the titanium liner 26 on the anolyte side. However, since nickel and steel are weldably compatible, there is no need to have a metal intermediate or wafer situated between them. This is not to say, however, that this invention excludes

the use of weldably compatible metal wafers between the cathode bosses 20 and the catholyte liner 48 when there is an anolyte liner 26 connected to the anode bosses 18, whether connected through metal intermediates or not. A liner may be used on one side, on both sides, or on neither side of the unitary cell element 12.

An anolyte compartment inlet opening 56, an anolyte compartment oulet opening 50, a catholyte compartment inlet opening 56, and a catholyte compartment outlet opening (not shown) are cast in the body of the flange 16 in that part of the flange which communicates with their respective anolyte compartment 22 and catholyte compartment 24. When there are liners 26, 48 in these compartments, then corresponding openings are provided in the liners. These openings can be seen in Figs. 1 and 2.

Conduits connected to the respective openings are shown in Figs. 4 and 5 as anolyte inlet conduit 60, anolyte outlet conduit 62, catholyte inlet conduit 64, and catholyte conduit 66.

Besides ferrous materials such as iron, steel, and stainless steel, cell element 12 can also be cast from any other castable metal such as nickel, aluminum, copper, chromium, magnesium, titanium, tantalum, cadmium, zirconium, lead, zinc, vanadium, tungsten, iridium, rhodium, cobalt, and their alloys. Catholyte side liners 48 are usually chosen from these materials also, with the general exception of magnesium and aluminum.

The anolyte and catholyte side liners 26 and 48 are preferably made of sufficiently workable metallic materials so as to be capable of forming into a single sheet and into the shape in which they are shown in the drawing. This includes the ability to be pressed so that they have frustoconically shaped caps 32 and 70. It should also be understood that the invention is not limited to the caps 32, 70 being frustoconically shaped nor limited to the anode and cathode bosses 18 and 20 being frustoconically shaped. They can be shaped and located so as to direct the flow of electrolytes and gas within the compartments 22 and 24. Bosses 18 and 20 should have their ends 28 and 40 flat and parallel with the flat electrode surface to which they are going to be connected. The ends 28 and 40 of the bosses 18 and 20 should present sufficient surface area to which electrical connections can be made to their respective electrodes to provide an electrical path with sufficiently low electrical resistance. The bosses 18 and 20 should be spaced so that they provide a fairly uniform and fairly low electrical potential gradient across the face of the electrode to which they are attached. They should be spaced so that they allow free electrolyte circulation from any unoccupied point within their respective electrolyte compartment to any other unoccupied point within that compartment. Thus the bosses will be fairly uniformly spaced apart from one another in their respective compartments. It should be noted here that although anode bosses 18 and cathode bosses 20 are shown in a back to back relationship across central barrier 14, they need not be. They can be offset from each other across barrier 14.

The materials from which anode and cathode bosses 18 and 20 are made are, of course, the same as that of the cell element 12 since part of this invention is to make them as an integral part of that cell element.

The anolyte and catholyte side liners 26 and 48 are required to be electrically conductive, resistant to chemical attack from the electrolyte compartment environment to which they are exposed, and sufficiently ductile to form the indented caps 32, 70.

Of course, the metals from which the anolyte and catholyte side liners 26 and 48 are made are usually different because of the different electrolytic corrosion conditions to which they are exposed. This is true not only in chlor--alkali cell electrolytes, but also in other electrolytes. Thus, the metals chosen must be chosen to fit the conditions to which they are going to be exposed. Typically, titanium is the preferred metal for the anolyte compartment liner 26. Other metals suitable for such conditions can usually be found in the following group: titanium, tantalum, niobium, hafnium, zirconium and their alloys.

The number of metals suitable for the catholyte side liner 48 is usually much larger than the number suitable for the anolyte compartment side principally due to the fact that most metals are immune from chemical attack under the relatively high pH conditions present in the catholyte and

due to the electrical cathodic protection provided by the metal on the anolyte side of the cell structure 12. Ferrous materials are usually preferred as the metals for the catholyte side liner, including steel and stainless steel. Other liner materials include nickel, chromium, tantalum, cadmium, zirconium, lead, zinc, vanadium, tungsten, iridium, cobalt and alloys of each of these metals.

As a general rule, the metal which is used for catholyte side liner 48 is also suitable for use in making the cathode 46. This is similarly true for the metal of the anolyte side liner 26 and its anode 36.

When a liner metal is used which is weldably incompatible with the metal of the cell structure 12, and when the liner 26 or 48 is to be connected to the cell structure 12 by welding, then metal intermediates or wafers are positioned between the cell structure bosses and the metal liners at the location where the welds are to be made. These may be in the form of a single metal wafer, in the form of a multilayered metal wafer, or in the form of a metal film formed either on the cell structure 12 or the liner 26 or 48.

<u>Example 1</u>

A cell structure specimen was cast of SA-216 grade WCB steel. The thickness of the central barrier was approximately 1.27 cm (0.5 in.) thick. The base diameter of the frustoconical boss was 7.62 cm (3 in.) and the top diameter was 3.81 cm (1.5 in.). Overall dimensions of the

structure were approximately 40.64 x 50.8 cm (16 x 20 in.), with ten bosses located on each side (anode and cathode) and directly opposed. The end to end distance of the bosses was about 6.35 cm (2.5 in.).

The finished casting showed surfaces of excellent quality. Sections were cut for further examination. Internal voids in boss sections were minimal or non-existant. The cell structure quality was deemed well suited for bipolar electrode service.

<u>Example 2</u>

A cell structure specimen was cast of SA-216 grade WCB steel. This particular structure represented a corner section for the proposed cell designed. Overall dimensions for the structure were approximately 61 x 61 cm (24 x 24 in.) with the central barrier being 1.27 cm (0.5 in.) thick. The base diameter of the frustoconical bosses was 7.62 cm (3 in.) and the top diameter was 3.81 cm (1.5 in.). The end to end distance of the bosses was about 6.35 cm (2.5 in.) as was the thickness of the periphery.

After casting, the specimen was machined on both anode and cathode sides so as to provide two parallel planes. The anolyte and catholyte peripheral structures were closely examined. No large voids and few small voids were found. The lateral faces of the periphery were suitable for finishing with a minimum amount of machine work necessary to meet gasketing and sealing requirements. Sections cut from the specimen revealed minimal or non-existant voids.

Example 3

Cell structures were cast for a nominal 1.22 m by 2.44 m (4 foot by 8 foot) electrolyzer press. The purpose of this example was to verify the castability of the particular shape and determine minimum central barrier thickness. The thickness of the central barrier of this structure was approximately 1.43 cm (9/16 in.). The base diameter of the frustroconical bosses was 7.62 cm (3 in.) and the top diameter was 3.81 cm (1.5 in.). The end to end distance of the bosses was about 6.35 cm (2.5 in.), as was the thickness of the periphery. The surfaces of the anode and cathode side were of acceptable quality with only minor surface imperfections present on the cope side of the casting. In repetitive use of the mold, no substantial variation in casting quality was observed. This example demonstrates that a steel casting of this size and shape was feasible for mass production of a cell structure.

CLAIMS:

1. A cell structure used in forming a bipolar electrode-type, filter press-type electrolytic cell unit, which unit is capable of being combined with other cell units to form a cell series; wherein in said series the cell structure is separated from adjacent cell structures by ion-exchange perm-selective membranes which are sealably disposed between each of the cell structures so as to form a plurality of cells; each of said cells having at least one membrane separating the anolyte and catholyte compartments of each cell; said cell structure having a central barrier which physically separates an anolyte compartment located on one side of the barrier from a catholyte compartment located on the opposite side of the barrier; said central barrier having an anode positioned in its adjacent anolyte compartment and a cathode positioned in its adjacent catholyte compartment; said central barrier having the anode of the adjacent anolyte compartment electrically connected through it to the cathode of the adjacent catholyte compartment; said anolyte and catholyte compartments which are adjacent to the central barrier having a peripheral structure around their periphery to complete the physical definition of said compartments; said cell structure also having an electrical current

conducting means associated with it for providing electrical current paths through the central barrier from its adjacent catholyte compartment to its adjacent anolyte compartment; and which cell structure includes anode and cathode stand--off means for maintaining the anode and cathode of the two electrolysis cells adjacent the central barrier at predetermined distances from the central barrier; characterized in that

the central barrier, the anode and cathode stand-off means, and at least part of the electrical current transfer means are integrally formed into a unitary central cell element made from a single casting of a castable material;

said anode stand-off means and that part of the electrical current connecting means located in the unitary central cell element on the anolyte side of the central barrier being combined into a multiplicity of anode bosses projecting a predetermined distance outwardly from the central barrier into the anolyte compartment adjacent the central barrier, said anode bosses being capable of being mechanically and electrically connected either directly to the anode of said anolyte compartment or indirectly to said anode through at least one compatible metal intermediate directly positioned between said anode and said anode bosses; and

said cathode stand-off means and that part of the electrical current connecting means located on the catholyte side of the central barrier being combined into a multiplicity of cathode bosses projecting a predetermined distance outwardly from the central barrier into the catholyte compartment adjacent the central barrier,

said cathode bosses being capable of being mechanically and electrically connected either directly to the cathode in said adjacent catholyte compartment or indirectly to the cathode through at least one compatible metal intermediate directly positioned between said cathode and said cathode bosses; and

said anode bosses being spaced apart in a fashion such that anolyte can freely circulate throughout the totality of the otherwise unoccupied adjacent anolyte compartment, and, likewise, said cathode bosses being spaced apart in a fashion such that catholyte can freely circulate throughout the totality of the otherwise unoccupied adjacent catholyte compartment.

2. The cell structure of Claim 1 wherein the castable metal of the unitary central cell element is a metal selected from iron, steel, stainless steel, nickel, aluminum, copper, chromium, magnesium, tantalum, cadmium, zirconium, lead, zinc, vanadium, tungsten, iridium, rhodium, cobalt, and alloys thereof.

3. The cell structure of Claim 1 or 2 including an anolyte side liner made of a metal sheet fitted over the surfaces on the anolyte compartment side of the cell structure;

said anolyte side liner being an electrically conductive metal which is resistant to corrosion due to the anolyte compartment environment;

said liner being formed so as to fit over and around the anode bosses and said liner

being connected to the unitary central cell element at the anode bosses; and

said liner being depressed sufficiently around the spaced anode bosses toward the central barrier in the spaces between the bosses so as to allow free circulation of the anolyte between the lined unitary central cell element and the membrane of the adjacent anolyte chamber, the liner replacing the unitary central cell element surface adjacent to the anolyte chamber as one boundary contacting the anolyte.

4. The cell structure of Claim 3 wherein the metal liner is connected to the anode bosses by welding through a metal intermediate which is disposed between the bosses and the liner, the metal of the metal intermediate being not only weldable itself, but also being weldably compatible with both the metal of the anolyte side liner and the metal of which the unitary central cell element is made.

5. The cell structure of Claim 3 wherein the unitary cell element is made of a ferrous material and wherein the anolyte side liner is made of a metal selected from titanium, tantalum, niobium, hafnium, zirconium, and alloys thereof.

6. The cell structure of Claim 5 wherein there are metal intermediates positioned between the anode bosses and the anolyte side liner, with each intermediate having at least two metal layers bonded together and with the outside metal layer of one side of the intermediate abutting the anode

boss and the outside metal layer of the opposite side of the intermediate abutting the anolyte side liner, the metal layer of the intermediate which abuts each anode boss being weldably compatible with the ferrous material of which the anode bosses are made and accordingly being welded to said anode bosses, and the metal layer of that side of the intermediate abutting the anolyte side liner being weldably compatible with the metal of which the anolyte side liner is made and accordingly being welded to said liner so that the liner is welded to the anode bosses through the intermediates.

7. The cell structure of Claim 5 wherein vanadium intermediates are interposed between the anode bosses and an adjacent titanium anolyte side liner, and wherein the titanium anolyte side liner is welded to the ferrous material bosses through vanadium intermediates.

8. The cell structure of Claim 1 or 2 including a catholyte side liner made of a metal sheet fitted over the surfaces of the unitary central cell element;

said catholyte side liner being an electrically conductive metal which is resistant to corrosion due to the catholyte compartment environment;

said liner being formed so as to fit over and around the cathode bosses and said liner being connected to the unitary cell element at the anode bosses; and

said liner being depressed sufficiently around the spaced cathode bosses toward the central

barrier in the spaces between the bosses so as to allow free circulation of the catholyte between the lined unitary central cell element and the membrane of the adjacent catholyte chamber, the liner replacing the unitary central cell element surface adjacent to the catholyte chamber as one boundary contacting the catholyte.

9. The cell structure of Claim 8 wherein the catholyte liner is connected to the cathode bosses by welding through a metal intermediate which is disposed between the bosses and the liner, the metal of the metal intermediate being not only weldable itself, but also being weldably compatible with both the metal of the catholyte side liner and the metal of which the unitary cell element is made.

10. The cell structure of Claim 8 or 9 wherein the unitary cell element is made of a ferrous material and wherein the catholyte side liner metal is selected from ferrous metals, nickel, nickel alloys, chromium, tantalum, cadmium, zirconium, lead, zinc, vanadium, tungsten, iridium, and cobalt.

11. The cell structure of Claim 8, 9 or 10 wherein there are metal intermediates positioned between the cathode bosses and the catholyte side liner, with each intermediate having at least two metal layers bonded together, the metal layer of the intermediate which abuts each cathode boss being weldably compatible with the ferrous material of which the anode bosses are made and accordingly being welded to said cathode bosses, and the metal layer of that side of the intermediate abutting the

catholyte side liner being weldably compatible with the metal of which the catholyte side liner is made and accordingly being welded to said liner so that the liner is welded to the cathode bosses through the intermediates.

12. The cell structure of Claim 8 wherein the metal of the unitary central cell element, of the catholyte side liner, and of the cathode of the adjacent electrolysis cell are all selected from ferrous metals.

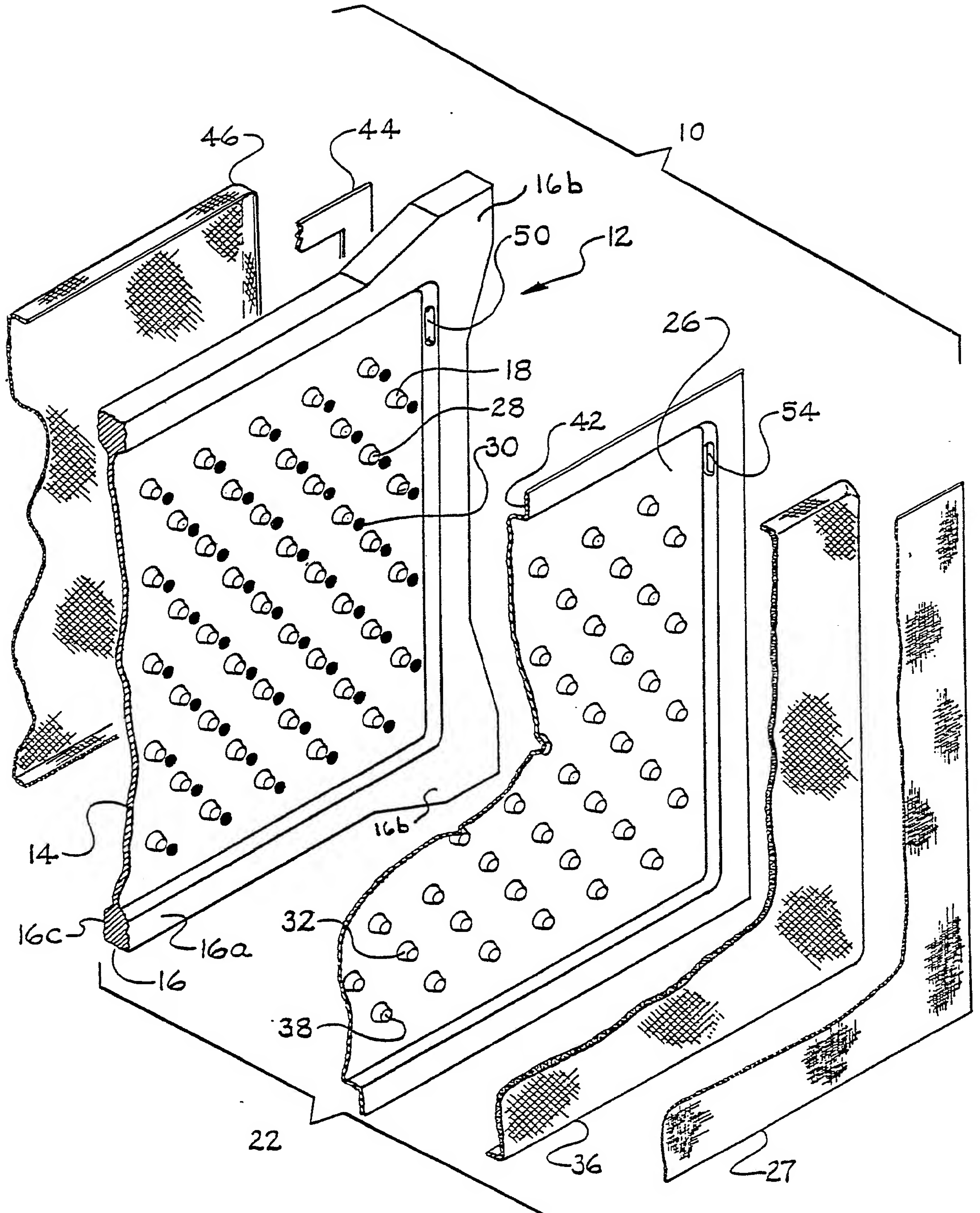
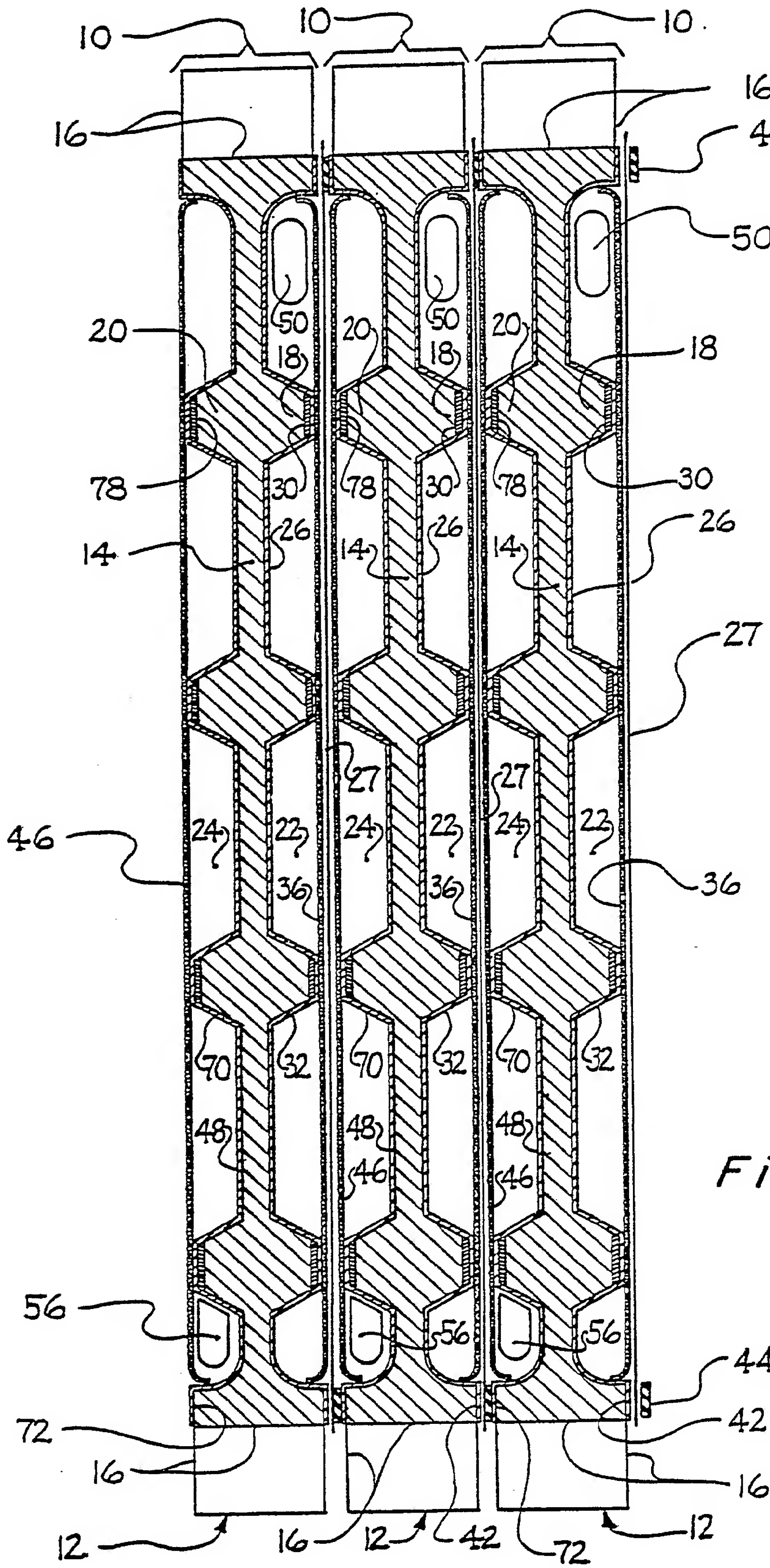


Fig. 1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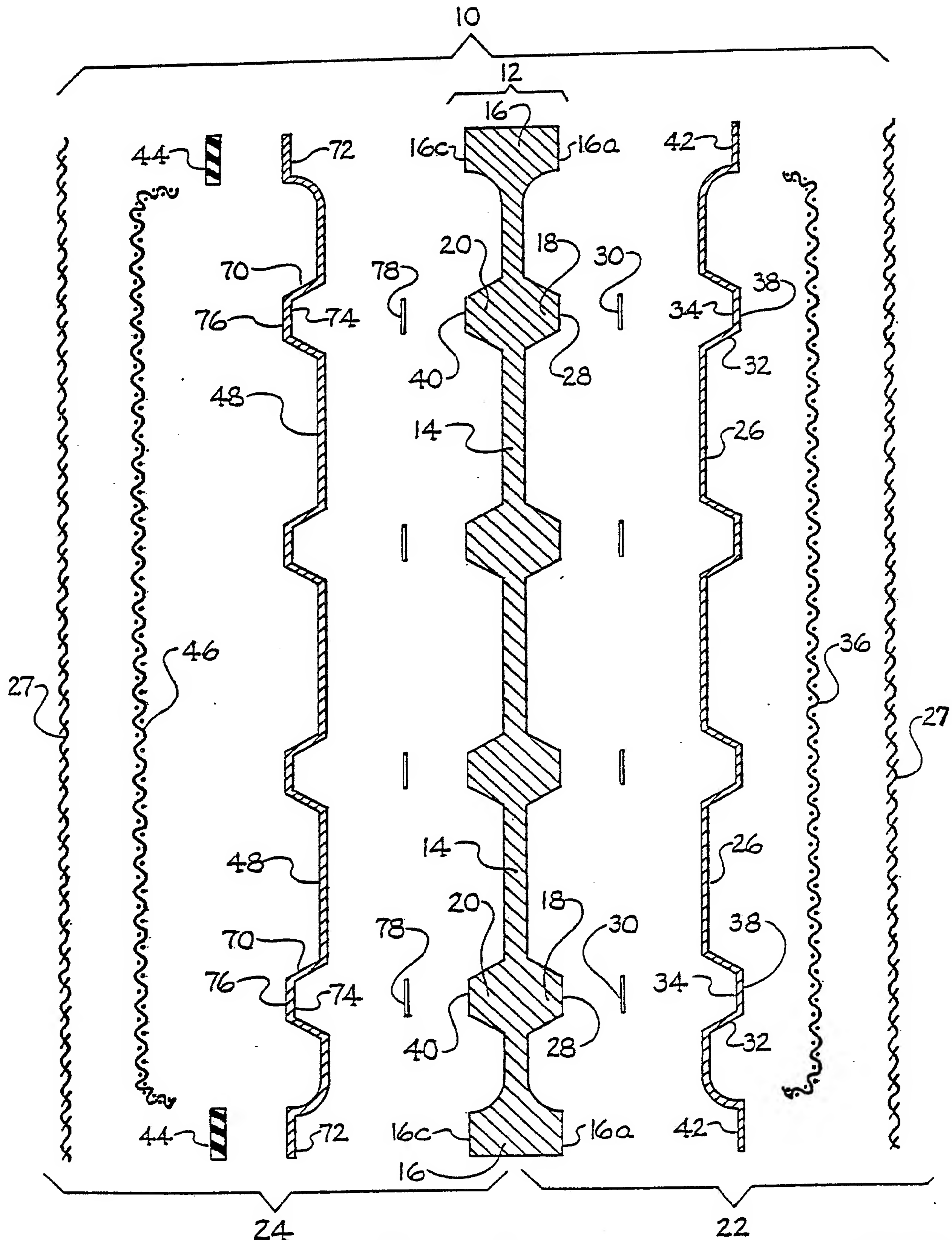
12

Fig. 2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16c
16a
42
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72
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38
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32
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14
26
46
36
27
27
14
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20
18
78
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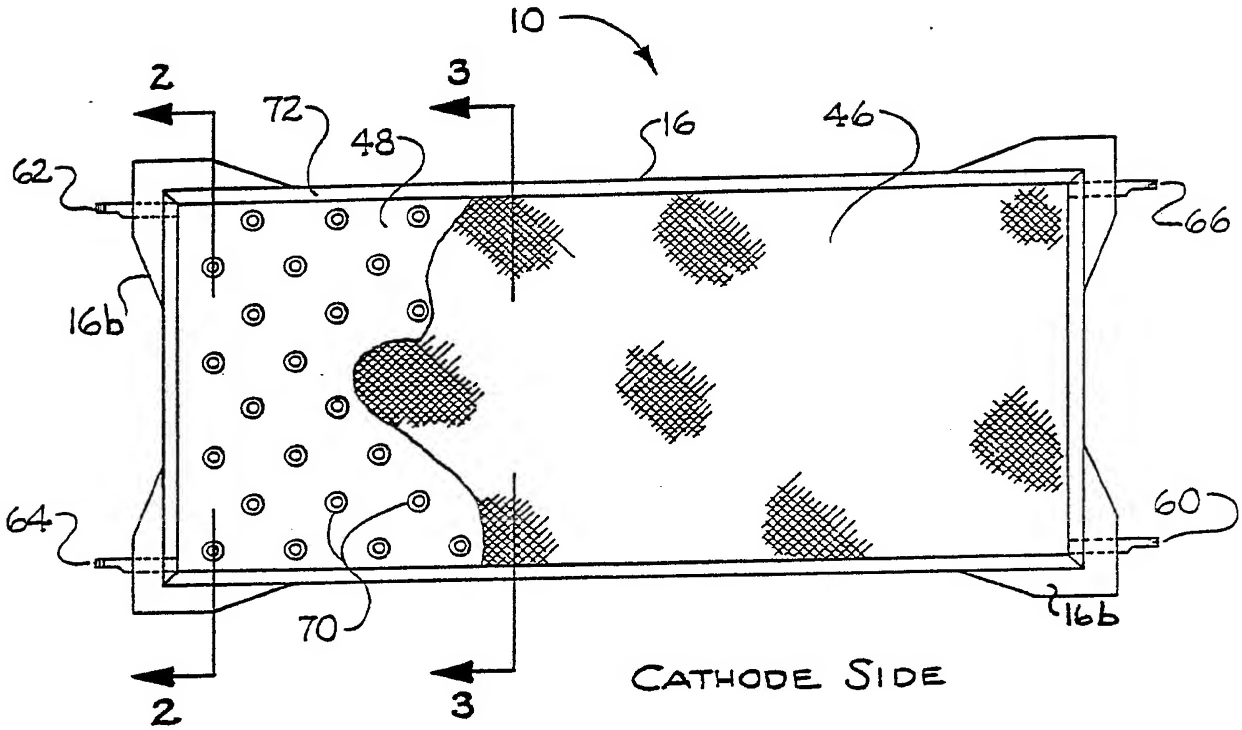
Fig. 3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2
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72
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16
46
62
66
16b
64
60
70
16b
2
3
CATHODE SIDE

Fig. 4

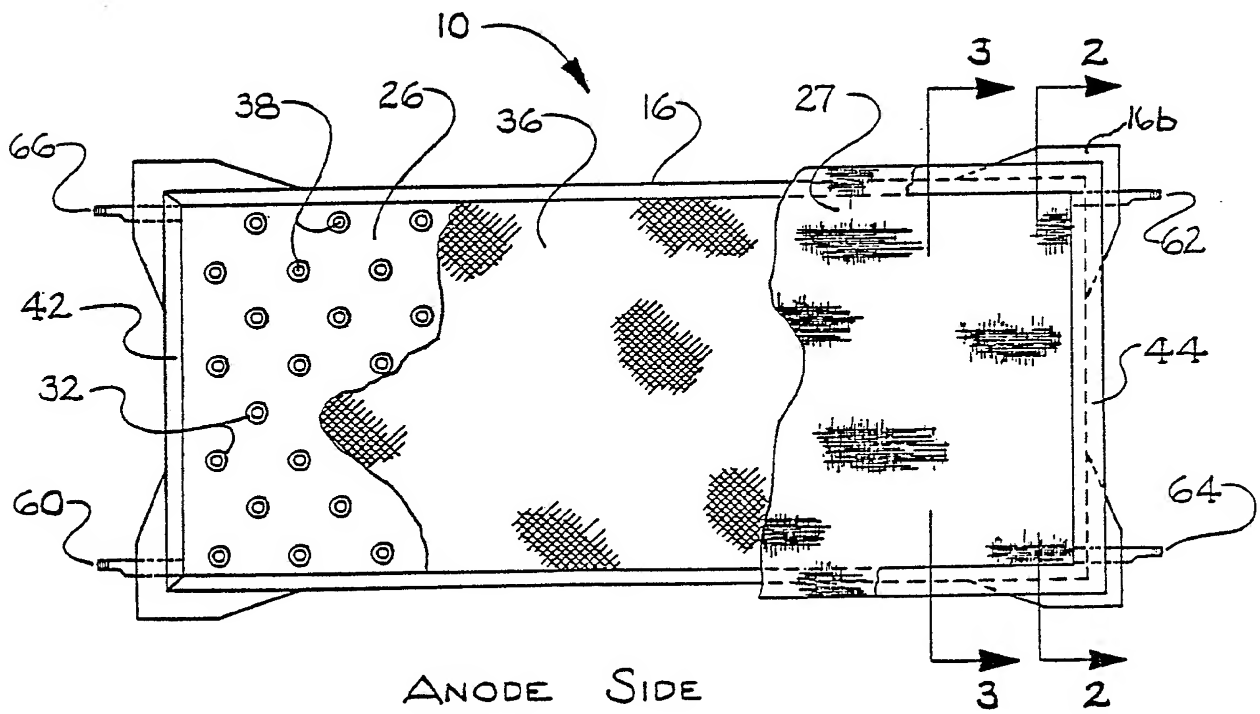
10
38
26
36
16
27
3
2
16b
66
62
42
44
32
60
64
3
2
ANODE SIDE

Fig. 5

INTERNATIONAL SEARCH REPORT

International Application No PCT/US84/00296

I. CLASSIFICATION OF SUBJECT MATTER (if several classification symbols apply, indicate all) [3]

According to International Patent Classification (IPC) or to both National Classification and IPC

INT. CL.[3] C 25 B 9/04
US CL. 204/254

II. FIELDS SEARCHED

Minimum Documentation Searched [4]

Classification System	Classification Symbols
US	204/254, 255, 256, 257, 258, 279, 286 and 297R

Documentation Searched other than Minimum Documentation to the Extent that such Documents are Included in the Fields Searched [5]

III. DOCUMENTS CONSIDERED TO BE RELEVANT [14]

Category *	Citation of Document, [16] with indication, where appropriate, of the relevant passages [17]	Relevant to Claim No. [18]
X	US, A, 3,960,698, (Bortak) 01 June 1976	1
A	US, A, 4,017,375, (Pohto) 12 April 1977	1-12
X	US, A, 4,197,178, (Pellegri et al) 08 April 1980	1
X	US, A, 4,214,969, (Lawrance) 29 July 1980	1
A	US, A, 4,279,731, (Pellegri) 21 July 1981	1-12
X	US, A, 4,339,322, (Balko et al) 13 July 1982	1

* Special categories of cited documents: [15]

"A" document defining the general state of the art which is not considered to be of particular relevance

"E" earlier document but published on or after the international filing date

"L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)

"O" document referring to an oral disclosure, use, exhibition or other means

"P" document published prior to the international filing date but later than the priority date claimed

"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention

"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step

"Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art.

"&" document member of the same patent family

IV. CERTIFICATION

Date of the Actual Completion of the International Search [2]	Date of Mailing of this International Search Report [2]
12 May 1984	22 MAY 1984
International Searching Authority [1]	**Signature of Authorized Officer [10]**
ISA/US	Terry Chapman Terry Chapman

Form PCT/ISA/210 (second sheet) (October 1981)